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Title: Distended GPHS Clads: Characterization of Fuel Particle Size

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GPHS Clads have been observed to distend or bulge slightly after aging or after storage in hot conditions. Configurations in which the fuel can reach a temperature above about 900°C appear to promote distension. [1] A distended clad is shown in Figure 1.



Figure 1. Image of distended GPHS clad FC0371

Alpha decay of the  $^{238}\text{PuO}_2$  GPHS fuel produces helium, which contributes to the distension as evidenced by the protruding vent cover seen in Figure 1. The vent cover is isolated from the fuel by a gas-permeable iridium frit. The frit assembly and the frit vent cover weld are shown in Figure 2.

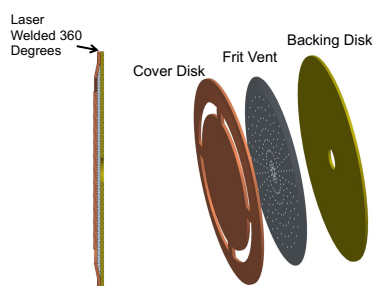


Figure 2. Detail of vent frit and exterior view of vent cover distended clad. The vent cover weld encircles the vent and vent cover assembly, isolating the frit from air.

The helium pressure which would be required in order to distend the iridium body of the clad is about 1500 psi, larger than the vent cover can sustain. [2] The vent cover would open before these large pressures could be achieved. Vent covers have been measured to give way at the weld under dynamic pressurization of pressures between 150 and 180 psig [3].

Fuel has been observed to have expanded from its original pellet size in radiographs of the distended clads, except for those clads in which the pellet is cracked. [1] Evaluation of the volume of the crack probably perturbs the estimation of volume in the cracked pellets, in units FCO371 and FCO435. Radiographs of FCO462 and FCO463 could not be scaled, and these units are omitted from the plot in Figure 3.

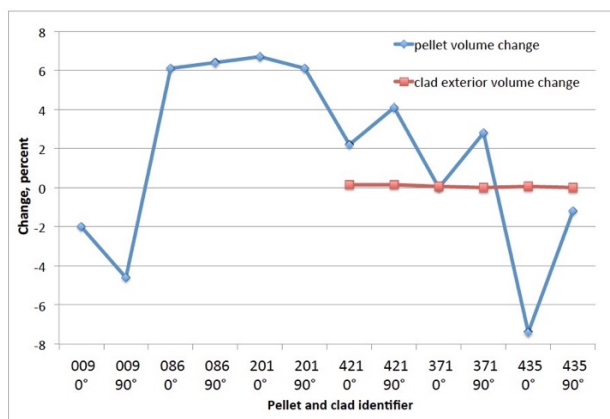


Figure 3. Pellet dimensions estimated from radiography

Radiographs of GPHS believed to have distended during normal storage.

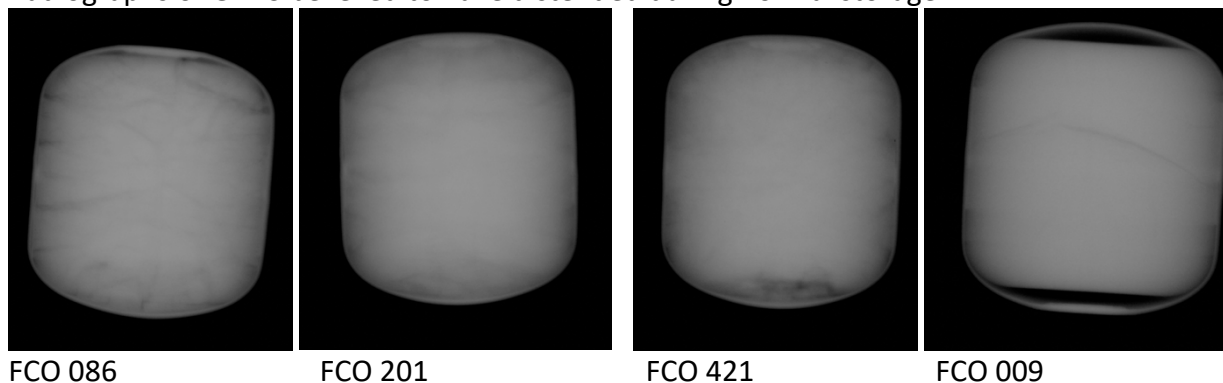
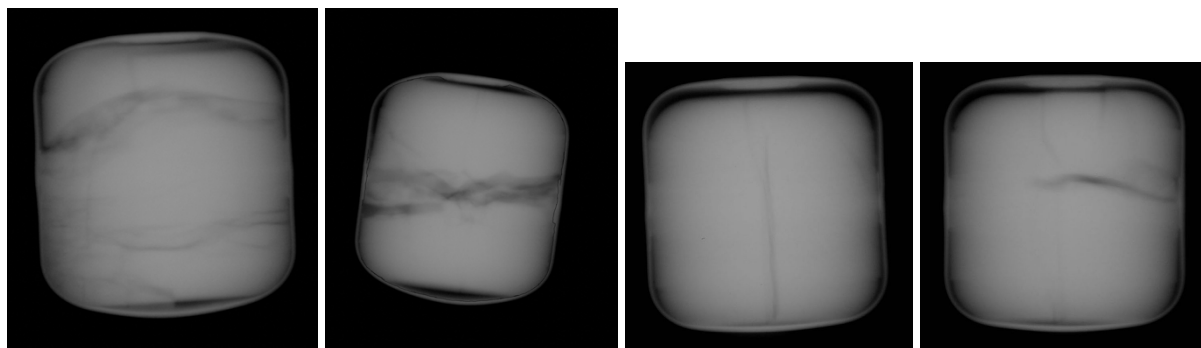


Figure 4. Radiographs of clads that became distended during normal storage. No radiography was performed on FCO312. Storage conditions encountered by FCO009 are not known.

Helium is known to be entrained in the fuel matrix [4] and in the pores incorporated into the fuel during pressing. This entrained helium may provide a mechanism for distension of the fuel. Fracture of the fuel in distended clads has also been observed, primarily in those clads that have suffered thermal excursions. Thermal strain may be the origin of the fracture, although abrupt release of helium from the fuel may also participate in or exacerbate the opening of cracks.

FCO009 contains substantial visible free volume, a characteristic which it shares with those units that have experienced thermal excursion, shown in Figure 5.



FCO 371

FCO 435

FCO 462

FCO 463

Figure 5. Radiographs of units known to have experienced high temperatures. No radiography was performed on FCO437.

Characterization of particle sizes in the fuel is a necessary component of examination and characterization of these distended clads. To this end, fuel from those distended clads which were previously examined and fuel from two recently discovered distended clads was sieved to establish fragment size distributions. All of the clads had been previously drilled to release any interior pressure. No pressure was observed in any clad examined.

Distension is an anomalous condition of GPHS clads that could be of concern to maintenance of the Safety Basis and assuring container integrity within the facility. Because no pressure has been found any distended clad, there is currently no perceived threat posed by distension of clads. All GPHS clads are stored singly in containers to prevent overheating, and containers are stored immersed in a cold water bath. Storage conditions for GPHS clads have been constrained to low temperatures to prevent thermal distension of GPHS clads, and clads are now required to have vent covers removed after 18 months, although the lack of pressure inside distended GPHS suggests that the vent cover spontaneously opens at some point in the GPHS life.

## Results of Measurement

Eight of the ten available distended clads have been defueled and the fuel sieved. Sieving data is shown in Figure 6. A graded series of 8 sieves are used, to obtain size fractions at 5600  $\mu\text{m}$ , 2000  $\mu\text{m}$ , 850  $\mu\text{m}$ , 425  $\mu\text{m}$ , 180  $\mu\text{m}$ , 125  $\mu\text{m}$ , 75  $\mu\text{m}$ , and 45  $\mu\text{m}$ , as described in procedure PA-DOP-01311, "Sieving of Plutonia." [5]

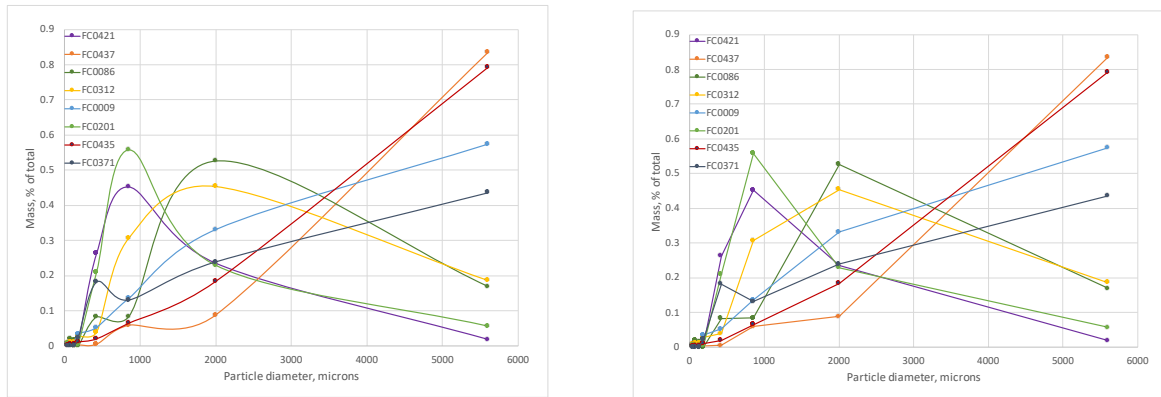


Figure 6. Fragment sizes in fuel from eight distended clads. Points represent measurements. Lines are drawn to guide the eye.

Clads examined to date fall into three groups. FCO201 and FCO421 exhibit the largest populations of particles in the 850  $\mu\text{m}$  size range, followed by FCO312. FCO086 and FCO312, show the largest populations of particles at 2000  $\mu\text{m}$ . FCO435 and FCO437 show the largest population of fragments larger than 5600  $\mu\text{m}$ , joined by FCO009 and FCO371. FCO435 and FCO371 show fractured pellets with large interior cracks in radiographs. FCO437 was not radiographed. FCO009 does not show evident interior cracks.

Units that have distended in thermal excursions, FCO371, FCO435, and FCO437, show similar particle size distributions with a predominance of large fragments, as shown in Figure 7. FCO009 is shown for comparison, and exhibits a similar distribution of fragment sizes.

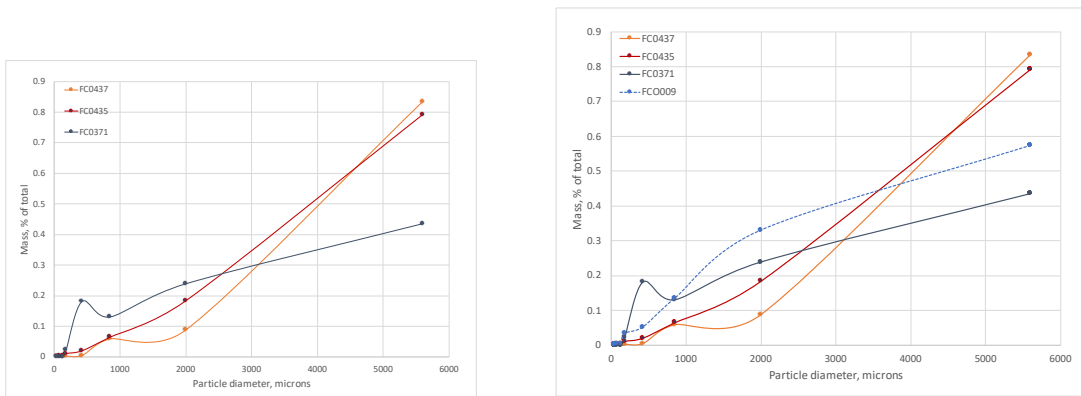


Figure 7. Particle sizes for clads exhibiting primarily large fragments. Clads shown in solid lines are known to have experienced thermal excursions

Large fragments in the fuel are consistent with large open cracks evident in the radiographs.

Conversely, aged units FCO421, FCO312, FCO201, and FCO086 show predominantly small particles, as anticipated from the uniformly cloudy radiographic images of the fuel. The unit FCO009 is the oldest clad in the inventory, but the fuel is more like a clad that has distended in a thermal excursion, as seen above. The history of FCO009 is not known, but it may have served as a neutron emission standard, and have been stored outside the cold water bath.

The population of particles smaller than 200  $\mu\text{m}$  does not increase with increasing age, although older clads appear to have somewhat higher populations of small particles than younger clads. FCO086 and FCO371 show the largest population, followed by FCO312. The oldest clad, FCO009, shows relatively few small particles.

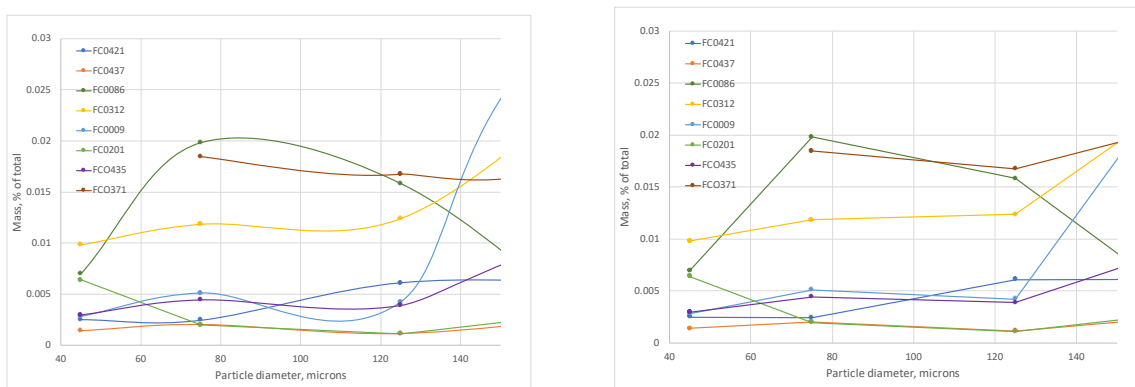


Figure 8. Fragment sizes smaller than 200  $\mu\text{m}$ .

Units with substantial populations of fines of 75  $\mu\text{m}$ , FCO086 and FCO312 are the same clads that show maxima around 2000  $\mu\text{m}$ , along with FCO009. FCO371 shows a substantial population at 75  $\mu\text{m}$ , but an intermediate population at 2000  $\mu\text{m}$ .

## Gas sampling

Pressures and analyses of the gas filling the distended clads indicates no pressure above atmospheric, only a small fraction of the anticipated helium inventory, and evidence of air inside the clad. These data suggest an open vent cover, and have been summarized elsewhere. [1] Recently discovered clads FCO462 and FCO463 have been recently sampled, and exhibit the same fill gas characteristic as previously examined clads. Data for these clads is summarized in Table 1.

**Table 1. Gas fill in FCO462 and FCO463**

Unit	Sampling date	Gas pressure	Gas composition
FCO462	14 May 2019	2.782 torr	includes air
FCO463	26 August 2019	0.951 torr	includes air

**Table 2. Distended Clads, a listing**

<u>Years</u>	<u>Clad</u>	<u>Fuel</u>	<u>distension mechanism</u>
6	FC0462	Russian	heat
6	FC0463	Russian	heat
9	FC0437	Russian	heat
9	FC0435	Russian	heat
12	FC0421	Russian	age 5.7 years at identification, 12 at gas sampling
17	FC0371	Russian	heat
23	FC0312	(Domestic)	age
24	FC0201	Domestic	age
25	FC0086	Domestic	age
26	FC0009	Domestic	age, possibly used as NER standard for some years

#### References

1. Roberta N. Mulford, Diane Spengler, Jonathan Teague, Jared Mason, Paul D. Richardson, Jack Gower, and Joey Sanchez, "Report on Distended GPHS Clads, Initial Measurements and Mechanism" Los Alamos National Laboratory Report LA-CP-16-20293 (May 2016)
2. J. Teague, "Integrity Evaluation of Swelled GPHS Clads" Los Alamos National Laboratory (15 October 2013)
3. J.P. Moore, "Quarterly Technical Progress Report of Radioisotope Thermoelectric Generator Materials Production and Technology Tasks for July through September 1995" Oak Ridge National Laboratory report ORNUCF-95/88 (October 20, 1995)
4. P. Angelini, R. E. McHenry, J. L. Scott, W. S. Ernst, Jr., and J. W. Prados, "Helium Release from  $^{238}\text{PuO}_2$  Microspheres," Oak Ridge National Laboratory Report ORNL-4507 (1970).
5. Los Alamos National Laboratory procedure "Sieving of Plutonia," PA-DOP-01311.